

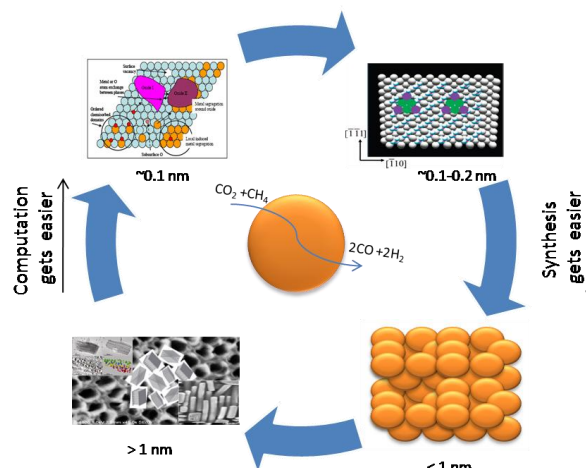


# Center for Atomic-Level Catalyst Design

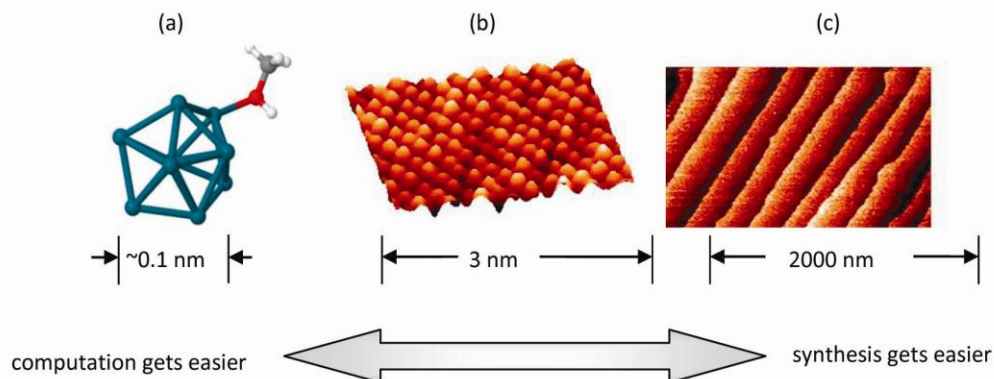
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**Mission Statement.** Our mission is to extend the capabilities of current computational and synthesis/ characterization tools and complement those tools with new approaches to atomic level synthesis and characterization of catalysts that achieve highest activity and selectivity in energy conversion processes.

**Rationale.** Recent advances in computational catalysis, and in our ability to synthesize materials with structural control at extremely small length scales, hold tremendous promise for dramatic improvements in catalysis. Despite rapid progress in both areas, we do not yet have the ability to (a) computationally design an ideal catalyst for a reaction of reasonable complexity at conditions of practical interest, nor to (b) prepare the catalyst with a degree of atomic-level precision that mimics the surfaces that we can simulate by computation.



This gap between simulated (computational) and real world catalysis can be visualized in a number of ways, e.g., by comparing the degree of difficulty in (a) calculating the interactions of even simple molecules with extremely small metal clusters and (b) in synthesizing these same clusters in a completely controllable way. Assuming other factors are constant (such as the complexity of the reaction), as the size of the active catalyst increases and the level of control over the atomic-level structure of the surface decreases, catalyst synthesis becomes easier—but the computational description of the catalyst (especially under reaction conditions) becomes less realistic—often because simplifying assumptions must be made that cannot be verified. Although the absolute length scale representing the limits of computation or synthesis may vary for different metals and reactions, the point is clear—there is a gap in our ability to identify an ideal catalyst by computation and to then prepare and characterize it unambiguously (Figure-1).



**Fig. 1. Difficulty of computational and synthesis/characterization research vary in opposite directions with length scale; (a)  $\text{CH}_3\text{OH}$  on  $\text{Rh}_7$  cluster (DFT model by D. Bruce, Clemson Univ.); (b)  $\text{Pt}_{25}\text{Rh}_{75}(100)$  Alloy 3-d topography ([www.omicron.de](http://www.omicron.de)); (c) Mono-atomic steps on  $\text{SiO}_2/\text{SiO}_2$  layer - in UHV at  $T = 800$  K in AFM mode.**

**Approach.** Our approach is two-fold; (a) extend the capabilities of current computational and synthesis/characterization tools and (b) complement those tools with newly developed approaches to synthesis and characterization. The computational effort will focus on developing first-principle based multi-scale models to predict catalytic behavior by following the dynamic evolution in both composition and structure over experimentally relevant time and length scales. The morphological changes and reactivity of the catalyst under various realistic conditions will be explored and the modeling predictions will be validated by experimental data. While Density Functional Theory (DFT) has been a very useful complement to surface science experiments, our proposed multi-scale modeling will be a step towards a more predictive role for computational simulations.

The synthesis effort is intended to extend--to ever-smaller dimensions--the length scales at which precise, computationally-specified structures of supported metals can be prepared. The approach will be an iterative process using synthesis and computation, in an integrated fashion. The proposed approach is anticipated to challenge the limits of both synthetic tools and computational techniques employed.

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